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**BORON CONCENTRATION GRADIENT FOR
IMPROVED THERMAL REACTOR PERFORMANCE
OF BORON-STAINLESS STEEL CONTROL RODS**

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CONTENTS

	<u>Page</u>
Abstract	1
Introduction	1
Description of the Concept	5
Theoretical Considerations and Mathematical Model	6
Design and Fabrication of Neutron Absorber Section Assembly . . .	10
Compatibility	13
Irradiation History	18
Postirradiation Examination	19
Visual Inspection	19
Burnup Analyses	20
Gamma Scanning	20
Chemical Analysis	20
Dimensional Measurements	23
Bond Integrity and Microstructural Effects	25
Discussion of Results	29
Conclusions	33
Acknowledgments	34
Appendix	
Width and Straightness Measurements	37
Plate Swelling Measurements	37

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ABSTRACT

The performance of conventional 2 to 3 wt % ^{10}B -stainless steel alloys and powder dispersions is limited in thermal reactors because the $^{10}\text{B}(n,\alpha)$ reactions are localized at and near the surface of the material. Significant swelling occurs after a relatively short exposure, and additional exposure drastically damages the microstructure in the localized regions. A symmetrical boron concentration gradient in which the ^{10}B is low at the surface and increases in a prescribed manner to higher concentrations below the surface substantially increased the performance capability of boron-stainless steel neutron absorbers.

This method was exploited in the 10-MW SM-1 Reactor. The neutron absorber section assembly of the control rod is a rectangular parallelepiped consisting of four 0.090-in.-thick plates clad with 0.032-in.-thick austenitic stainless steel. The ^{10}B concentration was set as 1 wt % ^{10}B at the surface of the plate and in a programmed manner increased to 3 wt % ^{10}B 0.024 in. below the surface. For an intended fluence of 4.3×10^{20} neutrons/cm² (thermal) this specific boron concentration gradient extends the ^{10}B consumption without damaging effects in any volume increment to 36×10^{20} atoms/cm³ (maximum). Thus, the allowable average ^{10}B burnup for the onset of swelling can be increased from 5 at. %, the limit for conventional dispersions, to 20 at. %. Postirradiation examination of a boron-gradient neutron absorber, which contained ^{10}B dispersed in type 200 austenitic stainless steel and was used for six years in the central region of the active lattice of this SM-1 Reactor, confirmed this improvement.

INTRODUCTION

Boron alloyed in stainless steel or dispersed in iron has limited application as neutron absorbers in thermal reactors. Conventional fabrication techniques for manufacturing these materials cause the boron to form intermetallic compounds with the iron, nickel, and chromium of the metal matrix. The result is a decrease in the ductility of these

materials.¹⁻³ Beard et al.⁴ found that boron-stainless steel alloys containing in excess of 2.5 wt % B tend to crack and are virtually impossible to fabricate by conventional methods.

Swelling caused by the $^{10}\text{B}(n,\alpha)$ reaction limits the service life of boron-metal dispersions. Richt et al.⁵ found that in 0.156-in.-thick stainless-steel-clad plates with a 0.090-in.-thick core containing 3 wt % B (92% enriched in ^{10}B), swelling initiates at an average ^{10}B burnup of only 5 at. % and the neutron damage progresses to severe destruction of the surface region at an average ^{10}B burnup of 16 at. %. This drastic effect is illustrated in Fig. 1 for a composite plate that swelled 6%. Approximately 25% of the neutron absorber has been extensively damaged, and obviously the ^{10}B burnup in this region is several times greater than the average. Such damage can be eliminated by a design that limits the burnup of ^{10}B atoms in each incremental layer through the plate to a constant acceptable value. These considerations result in the ^{10}B concentration gradient profile through the thickness illustrated in Fig. 2.

We tested this method by an experiment in one of the highest regions of thermal neutron flux in the 10-MW SM-1 Reactor. Our experimental absorber was a neutron absorber section assembly like the design specified for the Army's stationary medium-power reactors⁶ — namely, a

¹F. P. Ellen, *Mechanical Tests on Boron Steel*, RPD(c)/TM-207 (June 9, 1954).

²C. F. Leitten, Jr., R. J. Beaver, and A. E. Richt, "Stainless Steel Clad Dispersion of Boron in Iron for Pressurized Water Reactors," *J. Nucl. Mater.* 2: 136-44 (1959).

³J. R. Shettig, *Tensile and Impact Test Results on Irradiated Boron-Stainless Steel*, WAPD-SFR-FE-291 (Oct. 3, 1955).

⁴A. P. Beard et al., *Preparation of Nuclear Poisons and Control Alloys, Stainless Steel Base Alloys*, KAPL-1371 (June 17, 1955).

⁵A. E. Richt, C. F. Leitten, Jr., and R. J. Beaver, *Postirradiation Evaluation of Stainless Steel-Clad Plate-Type Specimens Containing 3 wt % Enriched Boron in Iron*, ORNL-TM-815 (June 1964).

⁶C. F. Leitten, Jr., R. J. Beaver, and J. E. Cunningham, *Specifications and Fabrication Procedures on Europium-Bearing Absorber Rods for Reactivity Control in Core II of SM-1*, ORNL-2733 (July 29, 1959).

Fig. 1. Typical Structure of a High-Burnup Fe- ^{10}B Miniature Absorber Specimen Showing Core-Cladding Separation. Etched: 2% picral. 100 $\times$.

rectangular parallelepiped whose sides are composite stainless-steel-clad plates in which the core and the cladding are nominally 0.090 and 0.032 in. thick, respectively. Testing this full-scale neutron absorber section assembly in the operating reactor offers the advantage of achieving a burnup profile from the tip end, where the burnup is highest, toward the opposite end, where the burnup is the lowest. Thus, at the termination of the test we can assess the ability of our absorber to withstand damage as a function of burnup of ^{10}B atoms. The term of irradiation was aimed at achieving near the tip end an average burnup of 20 at. %.

We chose type 200 austenitic stainless steel powder as a substitute for the iron powder previously used as the dispersant for the boron because of its higher corrosion resistance and its superior absorption

Fig. 2. Boron Gradient Designed to Achieve an Average ^{10}B Burnup of 20 at. %.

of epithermal neutrons compared to iron. We prepared the dispersions using powder metallurgy techniques for convenience in arranging a boron gradient concentration profile, and the fabrication procedures were essentially the same as established previously by Beaver et al.⁷

DESCRIPTION OF THE CONCEPT

The primary objective of the boron concentration gradient method is to distribute the $^{10}\text{B}(n,\alpha)$ reactions throughout as much of the neutron absorber as is feasible, with the concentrations of ^{10}B increasing from the surface inward to assure essentially the same burnup of ^{10}B atoms in any volume increment. The data reported by Richt et al.⁵ indicated that we could allow complete burnup of a dispersion of 1 wt % ^{10}B in iron (47×10^{20} atoms $^{10}\text{B}/\text{cm}^3$) without encountering swelling of stainless-steel-clad plates or undesirable damage to the metallurgical bond between the core of the plate and the cladding. We also limited the maximum concentration within the material to 3 wt % ^{10}B because this concentration is within existing technology. Thus, the concentration at the surface was established at 1 wt % ^{10}B , and the maximum concentration below the surface was limited to 3 wt % ^{10}B .

The intermediate concentrations (Fig. 2) are established with a computer program based on the theoretical burnup behavior of ^{10}B . The boron concentration profile is symmetrical for maximum service where the thermal neutron current penetrating each side of the absorber is essentially the same. The cross-hatched portions of the illustration represent the regions where the $^{10}\text{B}(n,\alpha)$ reactions principally occur. The vertical distance between the initial concentration profile and the profile after irradiation represents the ^{10}B burnup in any volume increment, in this design 36×10^{20} atoms $^{10}\text{B}/\text{cm}^3$ with a tolerance of $\pm 5\%$. The total cross-hatched area represents the average ^{10}B burnup anticipated and is 20 at. %.

⁷R. J. Beaver, R. C. Waugh, and C. F. Leitten, Jr., *Specifications for Army Power Package Reactor (APPP-1) Fuel and Control Rod Components*, ORNL-2225 (July 24, 1957).

THEORETICAL CONSIDERATIONS AND MATHEMATICAL MODEL

The principal objective of the boron gradient is to establish an initial concentration of ^{10}B in each finite increment of plate thickness to compensate for the decrease in neutron current from the surface inward. Theoretical considerations resulted in a rather complicated formula for the interior concentrations, and simplifying assumptions were resorted to.

Consider a parallel current of neutrons impinging perpendicularly on the surface of a layer. If S_i is the number of ^{10}B atoms/cm² of surface area in layer i , then the burnup rate of the ^{10}B atoms per unit area of surface layer is

$$\frac{dS_i(t)}{dt} = - \int_{x_i}^{x_{i+1}} I(x,t) N(x,t) \sigma dx, \quad (1)$$

where

x = distance through region i , cm

t = time, sec

$N(x,t)$ = ^{10}B atoms/cm³ at x and t in region i

$I(x,t)$ = neutron current, neutrons cm⁻² sec⁻¹

σ = microscopic cross section for the absorption of thermal neutrons by ^{10}B , cm²/nucleus.

Now consider space-averaged values of $I(x)$ and $N(x)$; σ is constant with both time and space. Then

$$\frac{dS_i(t)}{dt} = - I_i(t) N_i(t) \sigma \Delta x_i. \quad (2)$$

Since the rates of change of S_i and N_i are the same,

$$\frac{dN_i(t)}{dt} = - I_i(t) N_i(t) \sigma, \quad (3)$$

which has the solution

$$N_i(t) = N_i(0) \exp \left[- \int I_i(t) \sigma dt \right] . \quad (4)$$

The average number of ^{10}B atoms destroyed per unit volume is a measure of swelling and is the limiting quantity to be specified. It is calculated from

$$C_i(t') = N_i(0) - N_i(t') = N_i(0) \left[1 - \exp \left[- \int_0^{t'} I_i(t) \sigma dt \right] \right] , \quad (5)$$

where t' is the desired life of the rod.

The space-averaged neutron current in each region can be estimated as follows:

$$I_i(t) = I_{i-1}(t) \left[1 - N_{i-1}(t) \sigma \Delta x_{i-1} \right] \text{ for } i > 1 . \quad (6)$$

This model neglects scattering, absorption in anything but ^{10}B , changes in angular distribution of neutrons with x , and penetration of neutrons from the other side of the plate. Furthermore, it implies that one must know what the "perpendicular" neutron current is in region 1. To solve Eqs. (5) and (6) for $N_i(0)$, start with region 1 and assume that I_1 is independent of time and will be specified. Since $C_i(t')$ and t' can also be specified, $N_i(0)$ can be directly calculated: For region 1,

$$N_1(0) = C_1(t') / \left[1 - e^{-I_1 \sigma t'} \right] . \quad (7)$$

Substituting Eq. (7) into (4) gives

$$N_1(t) = \frac{C_1(t')}{1 - e^{-I_1 \sigma t'}} e^{-I_1 \sigma t} . \quad (8)$$

Substituting Eq. (8) into (6) gives

$$I_2(t) = I_1 \left[1 - \left(\frac{C_1(t') \sigma \Delta x_1}{1 - e^{-I_1 \sigma t'}} \right) e^{-I_1 \sigma t} \right]. \quad (9)$$

$N_2(0)$ is obtained by substituting Eq. (9) into (5) and integrating. This process of back-substitution is repeated to obtain all values of $N_i(0)$.

In the actual application, a numerical analysis was used in which t' was divided into very short steps Δt . In Eq. (5) the exponential term can be expanded and all but the first two terms discarded. Furthermore, a time-averaged value of I_i can be considered. The result is

$$\Delta C_i = N_i(0) \bar{I}_i \Delta t \sigma, \quad (10)$$

where $N_i(0)$ is the concentration at the beginning of each new time interval and

$$\bar{I}_i \Delta t = \bar{I}_1 \Delta t - \sum_{j=2}^i \Delta C_{j-1} \Delta x_{j-1}. \quad (11)$$

Substituting Eq. (11) into Eq. (10) gives

$$N_i(0) = \frac{\Delta C_i / \bar{I}_1 \Delta t \sigma}{\left[1 - \frac{\sum_{j=2}^i \Delta C_{j-1} \Delta x_{j-1}}{\bar{I}_1 \Delta t} \right]}. \quad (12)$$

One can specify $N_1(0)$ as the maximum permissible ^{10}B atoms destroyed, and therefore ΔC_1 for each of the time steps ($I_1 \Delta t$) can be calculated. Having established C_1 values, using the same time steps ($I_1 \Delta t$), and assuming a value for $N_2(0)$, one can calculate ΔC values for region 2. If the first trial is not successful, repetitions of this step will ultimately result in proper selection of the $N_2(0)$ value. The succeeding regions are similarly treated, and all initial concentrations can be established.

Our first trial at developing $N_i(0)$ from the described considerations was a relationship from which an average boron concentration gradient was approximated, as follows:

$$C_i(t') = \sum_0^{t'} \Delta C_i = \sum_0^{t'} N_i(0) \bar{I}_i \sigma \Delta t, \quad (13)$$

where the sum is over the time intervals Δt that t' is divided into. By considering average values of $N_i(0)$ and $\bar{I}_i$ over t' , Eq. (13) becomes

$$C_i(t') = \bar{N}_i(0) \bar{I}_i \sigma \sum_0^{t'} \Delta t, \quad (14)$$

where

$$\bar{I}_i t' = \bar{I}_1 t' - \sum_2^i C_{j-1}(t') \Delta x_{j-1}. \quad (15)$$

Substituting Eq. (15) into Eq. (14) yields

$$N_i(0) = \frac{C_i(t') / \bar{I}_1 t' \sigma}{\left[1 - \frac{\sum_2^i C_{j-1}(t') \Delta x_{j-1}}{\bar{I}_1 t'} \right]}. \quad (16)$$

As indicated in connection with the derivation of Eq. (6), these equations neglect scattering of the neutrons and thus overestimate the penetration of the neutron current. In our first application of the ^{10}B distribution technique, we roughly compensated for scattering by assuming that the perpendicular current was reduced by 1.3 times the number of absorptions. In addition to our basic requirement that $C_i(t')$ for each incremental region be equal and that each thickness increment be equal, we also assumed that $\bar{N}_1(0) = N_1(0)/2$. Thus, Eq. (16) becomes

$$\bar{N}_i(0) = \frac{1.3 \bar{N}_1(0)}{[1 - (i-1) \bar{N}_1(0) \Delta x \sigma]} \quad (17)$$

Having established this time-averaged distribution, we calculated the initial ^{10}B concentrations using successive short time steps, making corrections for changes in $\bar{I}_i$ and N_i at each time step. The total times used in the back-calculation to arrive at the initial concentrations were the times required to increase $\bar{N}_1(0)$ to $\bar{N}_1(0)$. It was assumed that all values of $N_i(0)$ occurred after the same lapse of time. Our results indicate that this method for approximating the $N_i(0)$ values for Eq. (12) produces a first-trial boron concentration gradient in which the ^{10}B atom burnup in each increment is reasonably constant.

DESIGN AND FABRICATION OF NEUTRON ABSORBER SECTION ASSEMBLY

Specific design details of the experimental neutron absorber section assembly are listed in Tables 1 and 2. The smooth boron gradient profile in Fig. 2 is not economically practical, but "staircasing" the profile is a reasonable and feasible approach. The "staircase" profile illustrated in Fig. 3 describes the actual layering of the specific ^{10}B concentrations and is symmetrical about the thickness midplane. The cross-hatched area describes the anticipated average burnup of 20 at. %, upon which the gradient is based. The total ^{10}B content of 9.36 g per plate complies with the "worth" requirements for the SM-1 control rod.

The fabrication practice consisted of mixing the exact quantities of elemental boron (with 92% enrichment in the ^{10}B isotope) and type 200 stainless steel powder (with no additive) for 2 hr, pressing in a die at 33 tons/in.², sintering in dry hydrogen at 1100°C, and coining in the same die at 33 tons/in.². The design of billets for encapsulating these compacts before rolling into composite plates provided for evacuation of the interior space. Figure 4 shows the arrangement of the components before encapsulation. The "barrier" foil in this case was used simply to assure a snug fit of the core. The rolling practice referenced previously for SM-1 neutron absorber plates was used;⁶

⁶C. F. Leitten, Jr., R. J. Beaver, and J. E. Cunningham, *Specifications and Fabrication Procedures on Europium-Bearing Absorber Rods for Reactivity Control in Core II of SM-1*, ORNL-2733 (July 29, 1959).

Table 1. Boron Gradient Profile of Experimental Neutron Absorber Assembly Core

Layer Type	Distance of Layer from Surface (in.)	Thickness of Layer (in.)	^{10}B Weight in Layer ^a (g)	^{10}B Concentration in Layer (wt %)
1	0 to 0.012	0.012	0.65	1.00
2	0.012 to 0.020	0.008	0.48	1.13
3	0.020 to 0.024	0.004	0.43	1.95
4	0.024 to 0.045	0.021	3.22	3.00
4	0.024 to 0.045	0.021	3.22	3.00
3	0.020 to 0.024	0.008	0.43	1.95
2	0.012 to 0.020	0.004	0.48	1.13
1	0 to 0.012	0.012	0.65	1.00
Total			9.56	

^aTotal B in layer: 10.34 g.

Table 2. Description of Experimental Neutron Absorber Assembly

Materials

Core of Plate: Elemental B, enriched to 92% ^{10}B , and type 200 stainless steel (0.03% C-20.70% Mn-16.37% Cr-0.49% Ni)

Frame and Cladding: Type 304L stainless steel (low cobalt)

Nominal Dimensions, in.

Composite Plate:	<u>Overall</u>	<u>Core</u>
Length	26.062	20.813
Width	2.464	2.214
Thickness	0.156	0.090

Length from end of plate to core end:

Tip end	0.250
Opposite end	5

Absorber Section Assembly:	<u>External</u>	<u>Internal</u>
Length	26.062	26.062
Width	2.620 × 2.620	2.308 × 2.308

Fig. 3. Actual Boron Gradient in Manufactured Composite Stainless Steel Plates for Experimental Neutron Absorber Assembly BG-1.

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Fig. 4. Components of the Billet for Manufacturing the Experimental Boron Gradient Neutron Absorber.

basically, the billet was reduced in thickness by a factor of 10.5. Preheating and reheating temperatures were 1100°C. The leading end of the plate was identified and was maintained as the lead end throughout the rolling operations. The objective of this schedule was to preserve as well as possible the boron gradient at this end, which is the end of the neutron absorber that receives the highest exposure. The rolling schedule gives these plates a minimum thickness at the end scheduled for the lowest ^{10}B burnup and a maximum at the end scheduled for the highest burnup. The actual measured dimensions of a representative plate are graphically illustrated in Fig. 5.

The plates were welded together to form the configuration illustrated in Fig. 6.

COMPATIBILITY

Our previous investigations had demonstrated the compatibility of elemental boron with iron,⁹ but the selection of austenitic stainless steel presented the question of the compatibility of the boron with the

⁹C. F. Leitten, Jr., R. J. Beaver, and A. E. Richt, "Stainless Steel Clad Dispersion of Boron in Iron for Pressurized Water Reactors," *J. Nucl. Mater.* 2: 136-44 (1959).

ALL DIMENSIONS ARE IN INCHES

Fig. 5. Critical Dimensions of Absorber Plate G-4.

Fig. 6. Locations of Dimensional Measurements of Welded BG-1 Neutron Absorber Section for SM-1 Test.

alloying elements at the elevated sintering and fabrication temperatures (1050 to 1150°C) normally used in manufacturing such absorbers.¹⁰ Austenitic stainless steel usually contains nickel to stabilize the austenite. However, the nickel-boron phase system has a eutectic¹¹ of 990°C at 42 at. % B. Our preliminary tests with intimate coupling

¹⁰C. F. Leitten, Jr., R. J. Beaver, and J. E. Cunningham, *Specifications and Fabrication Procedures on Europium-Bearing Absorber Rods for Reactivity Control in Core II of SM-1*, ORNL-2733 (July 29, 1959).

¹¹Max Hansen, *Constitution of Binary Alloys*, 2nd ed., p. 256, McGraw-Hill, New York, 1958.

of 4.6 wt % B and type 304 stainless steel showed evidence of melting after 30 min at 1050°C. Rather than limiting the processing temperatures to below 990°C, we selected a type 200 stainless steel in which the austenite is stabilized with manganese. Nickel in this alloy is a residual "tramp" element. The type 200 stainless steel tested contained 20.70 wt % Mn, 16.37 wt % Cr, 0.49 wt % Ni, 0.03 wt % C, 0.06 wt % Si, 0.008 wt % S, 0.009 wt % P, and the balance iron.

For compatibility testing we prepared 0.5-in.-diam specimens weighing nominally 7.32 g. Sets of samples were made to bracket the boron concentration we intended to use (1.15, 2.70, and 4.59 wt %) and also to bracket three possible sintering temperatures (1050, 1100, and 1150°C). The boron was 92% enriched in the ^{10}B isotope. Each sample was an individual entity containing the exact quantities of boron and stainless steel blended in a 1-oz bottle for 0.5 hr in an oblique blender and subsequently pressed under 33 tons/in.² into a green pellet. Each pellet was sintered at the selected temperature in a hydrogen atmosphere with a -60°F dewpoint, and subsequently coined under 33 tons/in.². Compatibility of coined samples was determined by visual examination for evidence of melting, and subsequently each sample was completely dissolved and chemically analyzed for boron. Several samples were encapsulated in type 304 stainless steel and rolled into composite plate specimens at the temperatures selected for sintering the specific compact. These clad specimens were examined metallographically for any evidence of incompatibility.

The results of the inspection of the coined pellets are in Table 3. No evidence of incompatibility was observed. The chemical analyses indicate that in general a slight loss of boron occurs. However, all results but one are well within the accuracy expected of the boron analyses at that time.

Metallographic examination of sections cut from the composite plate specimens confirmed the compatibility of these dispersions and indicated that in the range 1.15 to 4.59 wt % B no unusual problems should be encountered in producing full-size plates by the conventional rolling techniques. The compatibility of the dispersion containing 4.59 wt % B

Table 3. Compatibility of Elemental Boron^a with Type 200 Stainless Steel of Concentrations of 1.15 to 4.59 wt % at 1050 to 1150°C^b

Sample Identity	Sintering		Density (% of Theoretical)			Total Weight Loss (%)	Analyzed Boron (wt %)	Change from Boron Charged (%)
	Temperature (°C)	Time (min)	As pressed	Sintered	Coined			
1-3	1050	30	71	74	75	0.2	1.19	+3
7-9	1150	30	70	76	77	0.4	1.12	-3
19-21	1050	90	70	71	75	0.4	1.18	+3
28-30	1150	90	70	78	79	0.7	1.14	-1
37-39	1050	30	71	73	74	0.2	2.58	-4
46-48	1150	30	71	75	76	0.5	2.72	+1
55-57	1050	90	71	74	76	0.4	2.66	-2
64-66	1150	90	71	78	79	0.8	2.63	-3
73-75	1050	90	69	73	74	0.5	4.48	-2
82-84	1050	30	69	73	74	0.3	4.52	-2
91-93	1150	30	69	73	74	0.4	4.48	-2
100-102	1150	90	69	73	74	0.8	4.36	-5

^aEnriched to 92% ¹⁰B.

^bThe specimens were in excellent condition after sintering.

is worthy of emphasis. This result demonstrates that the fabrication limitations in producing stainless steel alloys containing concentrations in excess of 2.5 wt % B can be circumvented by the application of conventional powder metallurgical techniques and conventional roll-cladding processing in combination with the selection of type 200 stainless steel as the matrix.

We did not investigate the physical or mechanical properties of the fabricated dispersions. However, it is reasonable to assume that the structure in the finished product is primarily a mixture or solid solution of the borides of iron, manganese, and chromium dispersed in the type 200 stainless steel¹² and comparable in structure to the vacuum method boron-stainless steel alloys. Hardness traverses of dispersions showed that the hardness increased from 250 DPH for a 1.0 wt % ¹⁰B dispersion to 350 DPH in the 3 wt % ¹⁰B dispersion (hardness of the type 304 stainless steel cladding was 150 DPH). It would be expected that the tensile strength and elongation values are not significantly different than those reported for the boron-stainless steel alloys (18% elongation for 1.2 wt % B and 8% elongation for 2.0 wt % B).¹³

Corrosion was not tested, but this property of these boron dispersions in type 200 stainless steel would be expected to be comparable in pressurized water at 260°C with results reported for other boron-stainless steel alloys.¹⁴

IRRADIATION HISTORY

The experimental absorber section assembly, identified as BG-1, was charged into lattice position 44 (the central control rod location) of Core II of the SM-1 Reactor and operated as the neutron absorber

¹²Max Hansen, *Constitution of Binary Alloys*, 2nd ed., pp. 247, 249, 252, McGraw-Hill, New York, 1958.

¹³F. P. Ellen, *Mechanical Tests on Boron Steel*, RPD(c)/TM-207 (June 9, 1954).

¹⁴D. C. Belovin and U. Merten, *The Corrosion Behavior of Some Stainless Steel-Boron Alloys in High Temperature Water*, KAPL-UM-5 (Feb. 7, 1956).

of this control rod for the entire life of Core II (six years, one month). During this period the SM-1 Reactor produced 13.07 MWyr of thermal energy, and this neutron absorber was exposed to an estimated 1.2 effective full-power years of reactor service. The coolant was water at 230°C pressurized to 1200 psia, and the calculated center-line temperature of the neutron absorber was 290°C. The reactor coolant pH was maintained in the range of 8 to 10 by LiOH additions. Core II operations were characterized with numerous startup and shutdown cycles. We believe that the experimental absorber assembly was therefore subjected to a more severe test than would be contemplated under steady operating conditions.

POSTIRRADIATION EXAMINATION

The primary objective of this examination was to assess the dimensional stability and structural integrity of the absorber section assembly and its component plates after an exposure equivalent to an average ^{10}B burnup of 20 at. %. The program consisted of the following phases:

1. inspection of all surfaces visually and stereoscopically;
2. measurement of the critical dimensions of the assembly, including straightness, and measurement of the thickness of the individual plates;
3. inspection of the integrity of the cladding-to-core bonding by nondestructive ultrasonic techniques;
4. determination of the relative ^{10}B burnup within each plate by gamma scanning;
5. chemical determination of the average ^{10}B burnup within each plate; and
6. evaluation of the structural integrity of the boron-stainless steel dispersion by metallographic examination and by hardness measurements.

Visual Inspection

Scanning the surfaces of the neutron absorber section assembly with a low-power periscope and the in-cell stereomicroscope at magnifications to 20× failed to reveal any evidence of damage such as pits, cracks,

or blisters on any of the surfaces, including the weld surfaces, as a result of the irradiation.

A phenomenon believed to be related to exposure after removal from the reactor was observed. Rather than the black adherent oxide typical of other SM-1 absorber section assemblies examined previously in the hot cell, the oxide on this experimental component was grayish. Spectrographic analyses of this film indicated that in addition to the usual iron, nickel, and chromium corrosion products, significant quantities of calcium, silicon, zinc, and sodium were present. The source of these latter elements was not identified.

Burnup Analyses

Gamma Scanning

Stainless steel normally contains about 0.1 wt % ^{59}Co , which reacts to form ^{60}Co , which has a 5.3-year half-life. This provides a qualitative measure of the longitudinal ^{10}B burnup distribution, since it is related to the neutron current distribution. Gamma scanning was used to determine the ^{60}Co distribution; the output of a NaI scintillation detector was registered on a strip-chart recorder. Each plate was driven at a low constant speed past a collimating slit in the detector shielding. The collimator was $0.020 \times 1.00 \times 17$ in., and the detector was adjusted to record only gamma rays with energies greater than 1.0 MeV. One scan was made along the axial center line, and three scans were made across the width of each plate. The scans of each plate were essentially equivalent. A typical axial scan (plate G-4) is illustrated in Fig. 7. As anticipated, negligible ^{10}B burnup occurs in the upper 16 in. of the absorber.

Chemical Analysis

Sections were cut from plate G-4 in the locations illustrated in Fig. 8 for chemical analysis to determine burnup of the ^{10}B . This illustration also shows the location of sections cut for metallographic and hardness measurements. Lithium, as well as boron, was determined, and the average percent burnup of ^{10}B atoms was calculated:

Fig. 7. Axial Gamma Scan of Boron Gradient Neutron Absorber Plate G-4 from SM-1 Neutron Absorber Assembly BG-1.

Fig. 8. Sectioning Diagram for Plate G-4 from the SM-1 Absorber Element BG-1. Analytical burnup samples are designated as B-XX, metallographic specimens as M-XX, where XX indicates the distance from the upper end of the absorber plate.

$$\% \text{ BU} = \left(\frac{100}{0.92} \right) \left(\frac{{}^7\text{Li}}{{}^7\text{Li} + {}^{10}\text{B}} \right), \quad (18)$$

where

% BU = at. % burnup of ${}^{10}\text{B}$,

Li = wt Li after irradiation, and

B = wt B after irradiation.

Results are listed in Table 4. The 80 at. % value for sample B-25.25 is not considered comparable to the others because the core in this location was significantly thinner and therefore contained less ${}^{10}\text{B}$ than the other regions identified.

Table 4. Analytical ${}^{10}\text{B}$ Burnup Results on Sections From Plate G-4 of Irradiated Boron Gradient Section Assembly BG-1

Sample Identity ^a	Content (wt %)		Average ${}^{10}\text{B}$ Burnup (at. %)
	B	Li	
B-5.25	0.44	0.005	1.7
B-16.25	0.71	0.027	5.6
B-20.75	0.81	0.098	16.0
B-23.75	0.86	0.17	23.9
B-25.25	0.06	0.12	80.0

^aDistance in inches from upper end of neutron absorber plate.

Dimensional Measurements

Changes in the width and straightness of the irradiated neutron absorber assembly were detected by dimensional measurements along the assembly at the coordinates identified in Fig. 9 and compared to the preirradiation results. The procedures and apparatus are described in the Appendix. Table 5 summarizes the detailed results. A slight outward bowing occurred and increased with ${}^{10}\text{B}$ burnup to a maximum of 1.33%

Fig. 9. Coordinates for Width and Straightness Measurements on SM-1 Absorber Element BG-1. Element is shown as if viewed from the upper end of the assembly. Dimensions are in inches.

Table 5. Percentage Increase in Width of Absorber Section Assembly BG-1 After Irradiation Test

Distance from Upper End (in.)	Increase in Width, % for Each Measurement Row					
	1	2	3	4	5	6
10	0.04	0.04	0.00	0.22	0.17	0.13
14	0.13	0.27	0.09	0.48	0.40	0.31
18	0.31	0.22	0.35	0.44	0.35	0.27
22	0.44	0.22	0.35	0.93	0.93	0.84
24	0.53	0.40	0.66	1.33	1.33	1.20

in the region where the ^{10}B burnup was 24 at. %, but the assembly was comfortably within the maximum 2.650×2.650 in. boundary allowed by the reactor design.

Swelling of the component plates of the assembly was detected by measuring the irradiated assembly with procedures and equipment described in the Appendix and comparing the results with the preirradiation thickness measurements. These comparisons are also listed in detail in the Appendix. The results obtained on Plate G-4, believed to have had the highest neutron exposure, are illustrated in Fig. 10. Considering the uncertainties of measurement, the results indicate that swelling below a ^{10}B burnup of 16 at. % is negligible. Above this threshold, the swelling increases to 1.6% at a burnup of 23.9 at. %.

Bond Integrity and Microstructural Effects

Each of the four plates was inspected in its entirety with a Vidigage ultrasonic flaw detector. All tests revealed that the bond between core and cladding was intact. The only change in signal observed was in the tip end of each plate and is attributed to microcracking observed in this high-burnup region.

Plate G-4 was considered typical of the plates inspected. Transverse samples were cut from this plate in the locations illustrated in Fig. 8. The samples were mounted in epoxy resin, ground on successively finer grades of silicon carbide abrasive papers, polished in two stages on Syntron vibratory polishers using diamond polishing compounds, and examined both as polished and etched.

Hardness was measured with a Vickers hardness tester, and the results are illustrated in Fig. 11. They are related to the specific boron composition in the gradient as well as to the average ^{10}B at. % burnup. The embrittling effect reported previously for boron-stainless steel alloys is obvious but is not unexpected in view of previously reported results for boron-bearing dispersions, nor is the increase in hardness of the irradiated cladding surprising.

The nondestructive ultrasonic results for bond integrity were confirmed by the metallographic examination illustrated in Fig. 12,

ORNL-DWG 71-12402

Fig. 10. Comparison of Pre- and Postirradiation Thickness Measurements on Plate G-4.

Fig. 11. Hardness of Various Layers in Boron-Gradient Absorber Plates of Absorber Section Assembly BG-1 as a Function of Average Burnup of ^{10}B Atoms.

Fig. 12. Transverse Sections of Regions of Plate BG-4. As polished, 50 $\times$. Burnups of ^{10}B (a) 16 and (b) 24 at. %.

which clearly shows the integrity of the cladding-core bonding at ^{10}B burnups up to 24 at. %. The characteristics of the microstructure of the boron gradient after an average ^{10}B burnup of 16 at. % is shown in Fig. 13. The porosity present is generally smaller than 0.0004 in. and is typical of the preirradiated powder-metallurgically prepared gradient.

Figure 14 illustrates microcracking observed on one side of Plate G-4 in the region of 24 at. % burnup. The opposite side in this region was not cracked. Our ultrasonic inspections did not indicate the presence of these cracks, and we believe they formed when the samples were cut from the plate because of the extreme brittleness of this material.

The tip end high-burnup region is typified in Fig. 15. The boron gradient was obviously not preserved in this region. Therefore the cracks observed are not unexpected. Our postirradiation ultrasonic tests indicated that the integrity of the tip end region was questionable,

Fig. 13. Microstructural Features of the 1.0 wt % ^{10}B Region of the Boron Gradient Adjacent to the Cladding After a ^{10}B Burnup of 16 at. %. 500 $\times$. Etched with 2% picral.

and on this basis we believe the microcracks in this region existed before sectioning. The separation of the cladding from the frame in the region 0.125 in. beyond the end of the core is attributed to stringers, which were highly concentrated in ^{10}B and embrittled the cladding-core bond in this highly localized region.

DISCUSSION OF RESULTS

The primary objectives of the boron gradient were to (1) increase the resistance of the component to swelling and (2) eliminate the degradation of the metallurgical bond between the cladding and the boron-bearing core. Table 6 summarizes the results of our experiments and compares them with the performance of a conventional plate-type neutron absorber containing 3 wt % B (92% enriched) dispersed in iron. Whereas the conventional dispersion shows first signs of swelling at

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(a)

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(b)

Fig. 14. Microcracking Observed in One Side of Boron Gradient Plate After ^{10}B Burnup of 24 at. %. As polished. (a) 50 $\times$. (b) 500 $\times$.

Fig. 15. Typical Effects Observed at High-Burnup (50 at. % ^{10}B)
 Tip End of Plates. (a) Longitudinal section through tip end of plate G-1.
 6x. (b) Microcracking observed at tip end of plate G-4, 500x, etched.

Table 6. Comparison of Swelling in Plates Containing Boron Gradient with Swelling in Plates Containing Conventional Boron-Iron Dispersion

Average Burnup (at. %)	Thickness Change			
	Conventional ^a Fe-3% B		Boron Gradient ^b	
	(in.)	(%)	(in.)	(%)
4	none	0.0	none	zero
5	0.001	0.6	none	zero
12	0.003	1.9	none	zero
16	0.009	5.7	negligible	negligible
24	0.026	16.7	0.0025	1.62

^aApproximate surface temperature 80°C (ETR).

^bApproximate surface temperature 230°C (SM-1).

an average ^{10}B burnup of 5 at. %, swelling in the boron gradient is negligible at 16 at. % but becomes definite at 20 at. %. The most remarkable improvement however occurs at 24 at. % burnup, where the resistance to swelling has been increased tenfold!

To assure the integrity of the metallurgical bond between the boron-bearing core and the cladding, one of the base criteria of the gradient design was that the boron concentration in the neutron absorber adjacent to the cladding not exceed 1 wt % ^{10}B (47.38×10^{20} atoms $^{10}\text{B}/\text{cm}^3$). The metallographic results show that this bond between the cladding and material of this ^{10}B concentration is preserved at average burnups as high as 24 at. %.

The increase in hardness of the boron dispersion after irradiation was not unexpected, but the rate of increase in the 1.95 wt % ^{10}B and 3.00 wt % ^{10}B regions may be of interest to designers.

With respect to the performance of this absorber section assembly in the control rod of the SM-1 Reactor or in similarly designed thermal reactors, the results demonstrate the outstanding capability of a boron-gradient neutron absorber. After exposure to pressurized water at

1200 psia and 230°C during a six-year period, no deleterious corrosion effects of any significance occurred in the type 304 stainless steel cladding. Detailed dimensional measurements of the irradiated absorber section assembly after 13.07 MWyr reactor operation failed to reveal changes that adversely affected performance.

The only region of the component that we believe could have been significantly affected by the irradiation was the region within 0.5 in. of the tip end of the core, where we estimate the ^{10}B burnup exceeded 50 at. %. In this region the boron gradient was not preserved, and this fact could account for the damage and swelling observed. The data indicate that had the gradient been preserved, the swelling at this tip end would probably not have exceeded 2%. This defect can probably be corrected by refinements in fabrication practice.

CONCLUSIONS

1. Theoretical calculations demonstrate that the irradiation performance of boron-stainless steel control rods in thermal reactors can be improved by limiting the ^{10}B at. % burnup in each volume increment to a value below the threshold where microstructural damage and swelling become significant. This value is believed to be 47×10^{20} atoms/cm³ of ^{10}B (1 wt % B). This improvement can be accomplished by a boron concentration gradient in which the concentration is 1 wt % ^{10}B maximum at the surface and increases with depth in a programmed manner to 3 wt % ^{10}B maximum.

2. Stainless-steel-clad plate-type neutron absorbers with a 0.90 in.-thick boron concentration gradient reaching a maximum concentration of 3 wt % ^{10}B in the layer 0.024 in. below the surface can be manufactured by conventional powder metallurgy and hot roll bonding techniques, when type 200 stainless steel is selected as the matrix material for the ^{10}B .

3. Type 200 stainless steel and ^{10}B are compatible to concentrations to 4.59 wt % ^{10}B .

4. Swelling of the boron concentration gradient is insignificant after an average ^{10}B burnup of 20 at. % at an estimated center-line

temperature of 290°C. No microstructural damage is indicated. This conclusion represents a fourfold improvement compared to a conventional boron-iron dispersion containing 3 wt % ^{10}B .

5. Neutron absorber section assemblies containing a boron concentration gradient for use in the U.S. Army's 10-MW SM-1 Reactor can be manufactured to the required dimensional and control rod worth requirements using established techniques. Such an assembly is capable of withstanding exposures in this reactor to 1.2 full-power years without damage of any practical significance.

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APPENDIX

DETAILED POSTIRRADIATION DIMENSIONAL RESULTS OF BORON GRADIENT
PLATES AND NEUTRON ABSORBER SECTION ASSEMBLY BG-1

WIDTH AND STRAIGHTNESS MEASUREMENTS

The instrument shown in Fig. 16 consists of two opposed dial gage micrometers attached to the crossfeed mechanism of a modified metal-working lathe. The assembly is supported horizontally between the dial gages by brackets at each end. These support brackets are adjustable so that the assembly can be aligned parallel to the plane of the lathe bed. With the assembly properly aligned the extent of warp, bow, and twist in the assembly surfaces can be determined from the individual dial gage reading as the length or width of the assembly is traversed. The overall width of the assembly can be determined simultaneously from the sum of the dial gage readings once the instrument is properly calibrated.

Table 7 compares the assembly width measurements with preirradiation values. The complete trace of the postirradiation widths and outer plate profiles are shown in Fig. 17.

PLATE SWELLING MEASUREMENTS

The apparatus shown in Fig. 18 basically consists of a deep-throated (26-in.) caliper, which uses a precision roller bearing for the stationary anvil and a 0.0001-in.-indicating dial-gage micrometer for measurements. The caliper is attached to the left end of the support table by a pivot point and is counterbalanced so that the stationary anvil contacts the inner surface of the absorber plate with a low, constant force during the measurement. The support table is equipped with rollers so that the absorber element can be positioned easily for measurements of the plate thickness at any location along its length. A lifting mechanism is also provided to raise the caliper when the element is moved from one measurement location to the next. The equipment is calibrated periodically during the thickness measurements against precision standards and is thought to be accurate to within ± 0.0001 in.

The postirradiation thickness measurements on the four absorber plates are shown in Fig. 19. The thickness changes due to irradiation (swelling) are plotted in Fig. 20.

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Fig. 16. Equipment for External Width and Straightness Measurements.

Table 7. Comparison of Pre- and Postirradiation Width Measurements of SM-1 Absorber Section Assembly BG-1^a

Location ^b	Row No. 1			Row No. 2			Row No. 3		
	Pre-	Post	Change	Pre-	Post	Change	Pre-	Post	Change
2	2.613	2.613	0	2.616	2.609	-0.007	2.616	2.612	-0.004
6	2.619	2.617	-0.002	2.616	0	0	2.615	2.611	-0.004
10	2.617	2.618	+0.001	2.618	2.619	+0.001	2.616	2.616	0
14	2.618	2.621	+0.003	2.615	2.621	+0.006	2.615	2.617	+0.002
18	2.615	2.622	+0.007	2.619	2.624	+0.005	2.618	2.626	+0.008
22	2.615	2.625	+0.010	2.619	2.624	+0.005	2.618	2.626	+0.008
24	2.613	2.625	+0.012	2.620	2.629	+0.009	2.619	2.634	+0.015

Location ^b	Row No. 4			Row No. 5			Row No. 6		
	Pre-	Post	Change	Pre-	Post	Change	Pre-	Post	Change
2	2.619	2.628	+0.009	2.620	2.634	+0.014	2.612	2.625	+0.013
6	2.620	2.629	+0.009	2.618	2.629	+0.009	2.613	2.617	+0.004
10	2.620	2.625	+0.005	2.620	2.624	+0.004	2.613	2.616	+0.003
14	2.620	2.631	+0.011	2.621	2.630	+0.009	2.617	2.624	+0.007
18	2.620	2.680	+0.010	2.618	2.626	+0.008	2.614	2.620	+0.006
22	2.620	2.641	+0.021	2.620	2.641	+0.021	2.616	2.635	+0.019
24	2.620	2.650	+0.030	2.620	2.650	+0.030	2.613	2.640	+0.027

^aSpecification: 2.620 ± 0.004 in.

^bDistance from upper end of assembly in inches.

Fig. 17. Width and Profile Measurements Across SM-1 Absorber Section Assembly BG-1. Top: Across plates G-3 and G-5. Bottom: Across plates G-4 and G-6.

R-41795

Fig. 18. Equipment Used for Thickness Measurements of Irradiated Boron-Gradient Neutron Absorber Plates.

Fig. 19. Postirradiation Plate Thickness Results on Boron-Gradient Neutron Absorber Plates.

Fig. 20. Detailed Swelling Characteristics of Boron-Gradient Neutron Absorber Plates.